

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:
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Application No: 10/576,676

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For: BINDER FOR ELECTRIC
DOUBLE LAYER CAPACITOR
ELECTRODE

Examiner: PEZZUTO, HELEN LEE

DECLARATION PURSUANT TO 37 C.F.R. § 1.132

Commissioner for Patents
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Sir:

I, Takeshi OISHI, declare and say as follows:

1. I am familiar with U.S. Application Serial No. 10/576,676 as I am an applicant's co-worker.
2. The following experiment was conducted by myself.
3. This experiment demonstrates that the electrode and the electric double layer capacitor produced by using the binder of the present invention show high performance.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Takeshi Oishi

Signature

Takeshi OISHI

Typed or Printed Name

Oct. 5, 2010

Date

<Experimental report>

<OBJECT>

An object of this experimental report is to prove that the electrode with the binder of the present invention and the electric double layer capacitor with the electrode have extremely high performance.

<EXPERIMENT>

ADDITIONAL EXAMPLE 1

ADDITIONAL EXAMPLE 1 is corresponding to the Example 4 of the present application.

Into a reactor equipped with a stirrer were supplied 70 parts of ion exchange water, 0.2 part of sodium dodecylbenzenesulfonate, and 0.3 part of potassium peroxodisulfate, and then a gas part therein was purged with nitrogen gas. The temperature of the system was raised to 60°C. In another container, 50 parts of ion exchange water, 0.5 part of sodium dodecylbenzenesulfonate, 88.5 parts of butyl acrylate, 10 parts of acrylonitrile, 1 part of glycidyl methacrylate, and 0.5 part of 2-acrylamide-2-methylpropanesulfonic acid were mixed to yield a monomer mixture. This monomer mixture was continuously added to the above-mentioned reactor over 4 hours to conduct polymerization. During the addition, the reaction was conducted at 60°C. After the end of the addition, the inside of the system was stirred at 70°C for 3 hours, and then the reaction was ended. The polymerization conversion ratio was 98.5%. The composition ratio of the resultant polymer was consistent with the provided ratio of the monomers. The Tg of the resultant polymer is shown in Table 1. The reaction solution was cooled to 25°C and then thereto was added ammonia water to adjust the pH to 8. Thereafter, steam was introduced thereinto, so as to remove the monomers unreacted. Next, the resultant was concentrated to set the solid concentration to 40% so as to yield a binder composition A as a dispersion of an acrylate-based elastomer in water. This binder composition A was used to yield a polymer film. The tensile stress when the film was elongated at a ratio of 100%, and the elongation

when the film was cut were measured.

Next, a planetary mixer was used to mix 50 parts of acetylene black (powdery Denka Black, manufactured by Denki Kagaku Kogyo Kabushiki Kaisha) as an electroconductivity additive, 200 parts of a 5% carboxymethylcellulose solution in water (Cellogen 7A, manufactured by Daiichi Kogyo Seiyaku Co., Ltd.) as a dispersing agent, and 50 parts of water so as to disperse the electroconductivity additive, thereby yielding a dispersion of the electroconductivity additive, the solid concentration therein being 20%. An appropriate amount of water was added to 30 parts of the resultant dispersion, 100 parts of high-purity activated carbon powder having an average particle diameter of 5 μm and a specific surface area of 2000 m^2/g as an active material for an electrode, and 7.5 parts of the binder composition A, and then the resultant was mixed in a planetary mixer with appropriate amount of water to yield a slurry composition.

A doctor blade was used to apply the resultant slurry composition to an aluminum foil having a thickness 20 μm and the resultant was dried at 60°C for 20 minutes and at 120°C for 20 minutes, and subjected to roll pressing, thereby yielding an electrode having a thickness of 120 μm . The resultant electrode was heated at 150°C under a reduced pressure for 6 hours. The electrode density and the peel strength of the resultant electrode are shown in Table 1.

The electrode yielded as described above was cut into circles having a diameter of 12 mm. Electrode layer faces of these two electrodes were made opposite to each other, and a separator made of a rayon-based porous circular film of 16 mm in diameter and of 35 μm in thickness was sandwiched therebetween. This was put together with a spacer made of stainless steel into a coin-shaped outside container made of stainless steel (diameter: 20 mm, height: 1.8 mm, and thickness of the stainless steel: 0.25 mm). An electrolytic solution was incorporated therein without leaving air. Thereafter, the container was sealed up to produce an electric double layer capacitor. The used electrolytic solution was a solution wherein triethylmonomethylammonium tetrafluoroborate was dissolved in propylene carbonate at a concentration of 1.8

mol/L. The storage of the electrode after the heat-treatment and the fabrication of the capacitor were performed in a dry room having a dew-point temperature of -60°C . The electrostatic capacity and the internal resistance of the resultant electric double layer capacitor are shown in Table 1.

ADDITIONAL EXAMPLE 2

In ADDITIONAL EXAMPLE 2, 89.2 parts of butyl acrylate, 10 parts of acrylonitrile, 0.5 part of glycidyl methacrylate, and 0.3 part of 2-acrylamide-2-methylpropanesulfonic acid were mixed to yield a monomer mixture. Other than this, the polymerization process was carried out in the same manner as that of ADDITIONAL EXAMPLE 1 to obtain a polymer. The T_g of the resultant polymer is shown in Table 1. The reaction solution was cooled to 25°C and then thereto was added ammonia water to adjust the pH to 8. Thereafter, steam was introduced therein, so as to remove the monomers unreacted. Next, the resultant was concentrated to set the solid concentration to 40% so as to yield a binder composition B as a dispersion of an acrylate-based elastomer in water. This binder composition B was used to yield a polymer film. The tensile stress when the film was elongated at a ratio of 100%, and the elongation when the film was cut were measured. Then, other than using the binder composition B instead of using the composition A, the same process of above ADDITIONAL EXAMPLE 1 was applied in order to obtain the slurry composition, the electrode and the electric double layer capacitor, and evaluated the performance of them. The result is shown in Table 1.

ADDITIONAL EXAMPLE 3

In ADDITIONAL EXAMPLE 3, 86.5 parts of butyl acrylate, 10 parts of acrylonitrile, 3.0 part of glycidyl methacrylate, and 0.5 part of 2-acrylamide-2-methylpropanesulfonic acid were mixed to yield a monomer mixture. Other than this, the same polymerization process of above ADDITIONAL EXAMPLE 1 was applied thereby obtaining the polymer. The T_g of the resultant polymer is shown in Table 1. The reaction solution was cooled to 25°C and then thereto was added ammonia water to adjust the pH to 8.

Thereafter, steam was introduced thereinto, so as to remove the monomers unreacted. Next, the resultant was concentrated to set the solid concentration to 40% so as to yield a binder composition C as a dispersion of an acrylate-based elastomer in water. This binder composition C was used to yield a polymer film. The tensile stress when the film was elongated at a ratio of 100%, and the elongation when the film was cut were measured. Then, other than using the binder composition C instead of using the composition A, the same process of above ADDITIONAL EXAMPLE 1 was applied in order to obtain the slurry composition, the electrode and the electric double layer capacitor, and evaluated the performance of them. The result is shown in Table 1.

ADDITIONAL EXAMPLE 4

In ADDITIONAL EXAMPLE 4, 82.0 parts of butyl acrylate, 10 parts of acrylonitrile, 6.5 part of glycidyl methacrylate, and 1.5 part of 2-acrylamide-2-methylpropanesulfonic acid were mixed to yield a monomer mixture. Other than this, the same polymerization process of above ADDITIONAL EXAMPLE 1 was applied thereby obtaining the polymer. The Tg of the resultant polymer is shown in Table 1. The reaction solution was cooled to 25°C and then thereto was added ammonia water to adjust the pH to 8. Thereafter, steam was introduced thereinto, so as to remove the monomers unreacted. Next, the resultant was concentrated to set the solid concentration to 40% so as to yield a binder composition D as a dispersion of an acrylate-based elastomer in water. This binder composition D was used to yield a polymer film. The tensile stress when the film was elongated at a ratio of 100%, and the elongation when the film was cut were measured. Then, other than using the binder composition D instead of using the composition A, the same process of above ADDITIONAL EXAMPLE 1 was applied in order to obtain the slurry composition, the electrode and the electric double layer capacitor, and evaluated the performance of them. The result is shown in Table 1.

<RESULT>

(Table 1)

	ADDITIONAL EXAMPLE 1	ADDITIONAL EXAMPLE 2	ADDITIONAL EXAMPLE 3	ADDITIONAL EXAMPLE 4
The amount (wt %) of crosslinkable functional group	1.5	0.8	3.5	8.0
$T_g(^{\circ}\text{C})$	-43	-44	-43	-41
Tensile stress(MPa) at the time of the 100% elongation	0.8	0.8	1.0	1.4
Elongation(%) at the time of the cutting	700	700	650	550
Electrode density(g/cm ³)	0.640	0.640	0.640	0.630
Peel strength(N/cm)	0.100	0.100	0.120	0.150
Electrostatic capacity(F/g)	61.0	61.0	62.0	60.5
Internal resistance(Ω F)	3.12	3.12	3.12	3.20

<DISCUSSION>

As shown in Table 1, it is clear that the binder of ADDITIONAL EXAMPLES 1-4 has desirable Flexibility. It is also clear that the electrode with the binder and the electric double layer capacitor having the electrode show extremely high performance, that is, high electrode density, high peel strength, high electrostatic capacity and low internal resistance.